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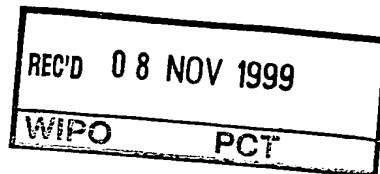
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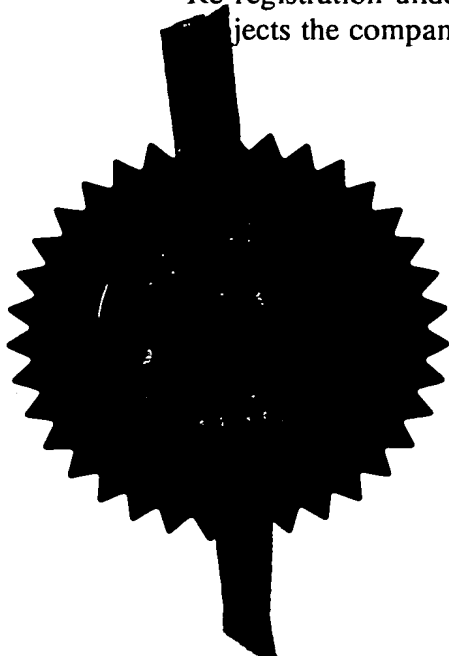
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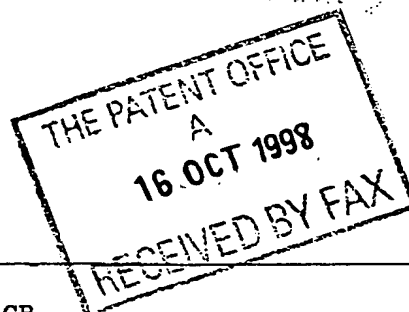
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1. Your reference

PFC 1430 GB

2. Patent application number

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16 OCT 1998

3. Full name, address and postcode of the or of each applicant (underline all surnames)

JOHNSON MATTHEY PUBLIC LIMITED COMPANY
2-4 COCKSPUR STREET
TRAFALGAR SQUARE
LONDON SW1Y 5BQ

Patents ADP number (if you know it)

If the applicant is a corporate body, give the country/state of its incorporation

J36268 007

4. Title of the invention

MEMBRANE

5. Name of your agent (if you have one)

LEONARD STUART BREWER

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

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 - b) there is an inventor who is not named as an applicant, or
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Signature L S Brewer Date 6 OCTOBER 1998
L S BREWER

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PFC 1430**DUPLICATE**

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MEMBRANE

The present invention relates to a solid polymer electrolyte and a membrane comprising the electrolyte that is of use in electrochemical devices, particularly fuel cells.

5 In particular, the invention relates to a process for the manufacture of the membrane electrolyte.

10 Electrochemical cells invariably comprise an ion-conducting electrolyte located between two electrodes, the anode and cathode, at which the desired electrochemical reactions take place. Electrochemical cells may be found in a range of devices, for example fuel cells, batteries, sensors, electrodialysis reactors and electrolytic reactors. They have a diverse range of applications, including the electrolysis of water, chemical synthesis, salt splitting, water purification, effluent treatment and metal finishing, among others.

15 A fuel cell is an energy conversion device that efficiently converts the stored chemical energy of its fuel and oxidant into electrical energy. It does so by combining either hydrogen, stored as a gas or methanol, stored as a liquid or a gas, with oxygen to generate electrical power. The hydrogen or methanol is oxidised at the anode and oxygen is reduced at the cathode. Both electrodes are of the gas diffusion type. The combined structure
20 formed from the membrane and the two gas diffusion electrodes is known as the membrane electrode assembly (MEA). An electric current is produced by the electron transfer reaction on the electrodes, with the flow of ions in the electrolyte completing the electrical circuit between the electrodes. The electrolyte therefore has to be in contact with both electrodes, and may be acidic or alkaline, and liquid or solid, in nature. In proton exchange membrane
25 fuel cells (PEMFC), the electrolyte is a solid, ion-conducting, *ie* a proton-conducting, polymer membrane. The movement of ions between the electrodes is dependent upon the presence of water in the membrane. The membrane must be fully humidified to provide the efficient movement of ions *ie* maintain the highest possible specific conductivity. The membrane also prevents large quantities of hydrogen gas supplied at the anode and oxygen
30 supplied at the cathode from passing through the electrolyte and mixing, although there is some transport of the reactant gases - a phenomenon referred to as "cross-over" - which leads to a reduction in cell performance. It is important that this cross-over is minimised. This is a particular problem with conventionally employed membranes at thicknesses below

around 40µm, which are otherwise favoured because of their high conductivity in the fuel cell.

Conventionally, the membrane electrolytes useful in fuel cells are selected from
5 commercially available membranes, for example the perfluorinated membranes sold under the trade names Nafion® (E I DuPont de Nemours and Co), Aciplex® (Asahi Chemical Industry) and Flemion® (Asahi Glass KK). In PEMFCs, the membrane electrolyte films - such as the Nafion® types of DuPont - have to be maintained in a highly water-saturated form in order to maximise their proton conductivity properties. Maintaining this high level
10 of water content under operating conditions requires a complex water management system.

Attempts to control the humidity of the membrane have been made, which include saturating the hydrogen gas supplied to the anode and air supplied to the cathode with water
15 vapour. However, this method brings with it its own disadvantages: it makes the overall system more complicated; lowers the fuel cell system efficiency; and can also reduce the cell's performance, particularly at the ability high current densities, due to increased reactant gas (particularly oxygen in air) diffusional problems as a result of the increased amount of water in the electrode layers.

20

It has been recognised that one way to overcome these difficulties is to employ the concept of a self-humidified membrane. One way that has been suggested that this can be achieved is through the use of water-retaining particles such as highly-dispersed silica (SiO₂) incorporated into the membrane electrolyte film. The membrane can retain a higher level
25 of water and is able to function with reduced need for humidifying the inlet gases.

It has also been discovered (European patent specification number EPA 631,337) that it is possible to incorporate catalyst materials, such as those based on platinum, into the particle containing membrane. This further improves the water retaining ability of the
30 membrane by reacting any hydrogen and oxygen that diffuse into the membrane from each of the catalysed electrode layers. This also can permit the use of thinner membranes, with

higher conductivity, by reducing the deleterious effects of reactant gas cross-over that would otherwise occur. Furthermore, the ability to use thinner membranes itself will further reduce the requirements for external humidification of the reactant gases.

5 The disclosure of EPA 631,337 relates to a solid polymer electrolyte composition comprising solid polymer electrolyte and 0.01 to 80% (based on weight of electrolyte) of at least one metal catalyst, and the use of this composition in fuel cells. The specification further discloses the optional incorporation of 0.01 to 50% of particles and/or fibres of one or more metal oxides into the electrolyte. However, the method disclosed for making such
10 compositions is multi-step and also has associated disadvantages. For example, one such composition is prepared by mixing a 5% w/w isopropanol solution of the electrolyte and an isopropanol dispersion of particles and/or fibres of 0.01 to 50% silica or titania and sufficiently agitating the mixture in an ultrasonic homogeniser. After this, the solution is transferred into a membrane moulding vessel and dried at an ordinary temperature, then
15 vacuum-dried at 60°C for removing the isopropanol so that the membrane is moulded. To incorporate the metal catalyst, in the case of platinum, the solid polymer electrolyte membrane containing the metal oxide is dipped in distilled water containing a platinum-ammine complex, followed by agitation at 60°C for 5 hours, during which the platinum complex replaces the protons on the membrane by ion-exchange and is adsorbed on the
20 membrane. After the membrane is washed with distilled water for over 4 hours, it is dipped in distilled water containing twenty times equivalence of hydrazine followed by agitation at 60°C for another 5 hours to deposit the platinum particles on the solid polymer electrolyte containing the metal oxide. Then, after the ion-exchange group is protonised by means of treatment with 4 molar concentration hydrochloric acid, the membrane is washed and dried.
25 Alternatively, the aforementioned EPA continues, after the metal catalyst such as platinum is supported on the metal oxide, the metal oxide may be incorporated in the solid polymer electrolyte in accordance with the same procedure.

30 Hence, it can be seen that the prior art process for forming the material is complex and involves multiple steps, many of which are time-consuming and may not be easily capable of producing a consistent material. These factors increase the cost of producing this

self-humidified membrane. It is essential for the future commercialisation of PEMFC power generation systems that the cost of the materials employed should be minimised. The polymer electrolyte membrane is one of the most expensive materials employed in the PEM fuel cell stack, due principally to the nature of the polymer itself, and its cost is currently too high to enable widespread commercialisation. Therefore, any process for producing more complex membranes needs to be simple, straightforward and low-cost.

A further expensive material in the PEM fuel cell stack is the platinum catalyst conventionally employed in the anode and cathode catalyst layers to promote the fuel cell electrode reactions of hydrogen oxidation and oxygen reduction. For many applications, particularly transportation, there is a need to minimise the use of these materials. It is widely believed that, for successful commercial exploitation, the level of platinum in the fuel cell stack should be reduced to a cell loading of as low as $0.20\text{mg}/\text{cm}^2$ of the MEA. Therefore, whilst the incorporation of an additional amount of platinum catalyst into the membrane to effect the combination of any reactant gases permeating into the membrane to produce an improved self-humidified membrane is a worthwhile objective, it is a requirement that this additional amount of platinum be as low as possible. EPA 631 337 and J Electrochem Soc 143 (12) 3847-52 (1996) indicate that platinum loadings of $0.07\text{mg}/\text{cm}^2$ are employed in the films produced by the above described multi-step process. At some 35% of the total platinum content that may be allowable for vehicular applications of the PEM fuel cell, this may be too high a loading for economic reasons. To our knowledge, therefore, membranes of this type have not yet found application in any of the current, practical pre-production demonstrations of PEM fuel cell power generators.

It is therefore an object of the present invention to overcome the disadvantages of the conventional ion-exchange membranes, without incurring the disadvantages of the prior art process for manufacturing a self-humidifying membrane.

Accordingly, the present invention provides a process for preparing a solid polymer electrolyte composition comprising an ion-conducting polymer, a catalyst and a high surface area support material, which process comprises:

5

- (a) associating the catalyst with the support material to form a catalysed support; and
- (b) combining the catalysed support with an aqueous ion-conducting polymer composition.

5 Preferably, the aqueous ion-conducting polymer composition is one disclosed in European patent specification number EPA 731 520, the contents of which are incorporated herein by reference in their entirety. Accordingly, the process of the present invention preferably comprises, as step (b), combining the catalysed particulate material with an ion-conducting polymer in a liquid medium that is aqueous-based and is essentially free from
10 organic solvents.

The process according to this invention, compared with the prior art multi-step process, is less complex, has fewer and shorter process steps, and is capable of producing a consistent and reproducible product; hence, the cost of producing the membrane is thereby
15 reduced. Furthermore, as the catalyst is directly applied to the high surface area particulate material, which thereby acts as the support for the catalyst, the catalyst is also deposited in a high surface area, small particle size form. This enables the expensive catalyst to be used more effectively and in lower amounts whilst also improving the self-humidifying performance of the membrane. Additionally, when the particulate material has water-
20 retaining properties, all of the catalyst promoting the recombination of hydrogen and oxygen is in immediate and intimate contact with the sites at which the water formed can be stored.

The catalyst may comprise any catalyst material capable of enabling the reaction of
25 hydrogen and oxygen to form water to occur at PEM fuel cell temperatures of typically below 150°C. These include the precious metals, platinum, palladium or rhodium, or combinations thereof, or other transition group metals such as nickel or cobalt. Preferably, the precious metals platinum, palladium or rhodium, or combinations thereof are employed and, more preferably, platinum is used. Suitable catalysation processes, as already practised
30 on a commercial scale, can be employed to form the catalyst material. These processes permit very close control of the amount of catalyst metal deposited onto the support, and

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thus subsequently incorporated into the membrane. Typically the catalyst metal can be deposited onto the support to loadings of between 0.01 to 50.0% by weight of the total catalyst, and more preferably from 1 to 25 wt%, such as, for example 10 wt%. The amount of catalyst material incorporated into the membrane would be such that if it were a precious
5 metal, the loading would be lower than 0.1mg/cm^2 , preferably lower than 0.05mg/cm^2 and, more preferably, lower than 0.03mg/cm^2 .

The high surface area support material may comprise any known, non-electrically conducting material which can be used as a support for a catalyst, such as silica, titania,
10 alumina, zirconium oxides, zirconium silicates, tungsten oxides, tin oxides and zeolites. It may also be advantageous to employ support materials that are known to retain high levels of water once suitably wetted, such as, for example, forms of silica. The material may be in the form of either fibres or small particles, such as particles with a mean particle size in the range of from $0.001\mu\text{m}$ to $10\mu\text{m}$, preferably $0.01\mu\text{m}$ to $5\mu\text{m}$.

15 For PEM fuel cell applications, the ion-conducting polymer is a proton-conducting polymer, examples of such polymers being well known to those skilled in the art. More than one proton-conducting polymer may be present and/or a non-proton-conducting polymer may also be included.

20 Proton-conducting polymers suitable for use in the present invention may include, but are not limited to:

1. Polymers having a substantially fluorinated, straight or branched carbon chain,
25 further having acidic groups or derivatives thereof selected from the following acids: sulphonic, carboxylic, phosphonic, phosphoric, and/or mixtures thereof. Perfluorinated polymers include Nafion[®], Flemion[®] and Aciplex[®], which are commercially available from E I DuPont de Nemours (cf US patents specifications numbers 3,282,875; 4,329,435; 4,330,654; 4,358,545; 4,417,969; 4,610,762; 4,433,082 and 5,094,995); Asahi Glass KK and
30 Asahi Chemical Industry, respectively. Other such polymers include those disclosed in US patents specifications numbers 5,595,676 and 4,940,525.

2. Perfluorinated or partially-fluorinated polymers further having aromatic rings, such as those described in PCT patents specifications numbers WO 95/08581, WO 95/08581 and WO 97/25369, which have been functionalised with SO_3H , PO_2H_2 , PO_3H_2 , $\text{CH}_2\text{PO}_3\text{H}_2$, COOH , OSO_3H , OPO_2H_2 , and/or OPO_3H_2 . Also included are radiation- or chemically-
5 grafted perfluorinated polymers, in which the perfluorinated carbon chain is activated by radiation or chemical initiation in the presence of a monomer, such as styrene, which can be functionalised to contain an ion-exchange group. Suitable perfluorinated polymers include, for example, PTFE, fluorinated ethylene-propylene (FEP), tetrafluoroethylene-ethylene (ETFE) copolymers, tetrafluoroethylene-perfluoroalkoxy (PFA) copolymers, poly (vinyl
10 fluoride) (PVF) and poly (vinylidene fluoride) (PVDF).

3. Fluorinated polymers, such as those disclosed in European patent specifications numbers EP 0 331 321 and EP 0345 964, having pendant saturated cyclic groups and at least one ion-exchange group that is linked to the polymeric chain through the cyclic group.

15 4. Aromatic polymers, such as those disclosed in European patent specification number EP 0 574 791 and US patent specification number 5 438 082, for example, sulphonated polyaryletherketones. Also polyether sulphones, which can be chemically grafted with a polymer with ion-exchange functionality, such as those disclosed in PCT patent specification
20 number WO 94/16002.

5. Non-fluorinated polymers, including those disclosed in US patent specification number 5 468 574, for example, hydrocarbons such as styrene-(ethylene-butylene)-styrene, styrene-(ethylene-propylene)-styrene and acrylonitrile-butadiene-styrene co- and ter-
25 polymers, where the styrene components are functionalised with sulphonate, phosphoric and/or phosphonic groups.

6. Nitrogen-containing polymers, including those disclosed in US patent specification number 5 599 639, for example, polybenzimidazole alkyl sulphonic acids and poly-
30 benzimidazole alkyl or aryl phosphonates.

7. Any of the above polymers wherein the ion-exchange group is replaced with a sulphonyl chloride (SO_2Cl) or sulphonyl fluoride (SO_2F) group, thereby rendering the polymers melt-processable. The sulphonyl fluoride polymers may form part of the precursors to the ion-exchange membrane or may be arrived at by subsequent modification of the ion-exchange membrane. The sulphonyl halide moieties can be converted to sulphonic acid using conventional techniques such as, for example, hydrolysis.

Other ion-conducting polymeric materials which are not proton-conducting may be used in addition to, or in place of, a proton-conducting polymer. Such polymers can be used for applications requiring a bipolar membrane or a completely anion exchange membrane. Anion exchange polymers are generally based on quaternary ammonium groups, rather than the fixed sulphonic acid groups in proton conducting polymers. These include, for example, the tetra-alkyl ammonium group ($-\text{N}^+\text{R}_3$) and the quaternary ammonium centre in Tosflex[®] membranes ($-\text{N}(\text{R}_1)(\text{CH}_2)_y\text{N}^+(\text{R}_3)$) supplied by Tosoh. However, it can be envisaged that all of the proton exchange polymers described above could have anion exchange equivalents.

Other non-ion-conducting polymeric materials may be used in addition to the one or more ion-conducting or proton-conducting polymers. Examples of such non-ion-conducting polymers include fluorinated polymers, eg PTFE, FEP, PVDF, Viton[®], and hydrocarbon types eg polyethylene, polypropylene and polymethylmethacralate.

Most preferably, the ion-conducting polymer is an essentially aqueous solution of a perfluorinated co-polymer with ion-exchange groups, such as Nafion[®] (as mentioned hereinbefore).

The solid polymer electrolyte composition prepared by the process of the present invention may be employed as or as a component of the ion-exchange membrane of a solid polymer electrolyte chemical cell, such as a PEMFC. Accordingly, the present invention further provides a membrane comprising a composition prepared by the process of the present invention.

The catalysed support can be incorporated, *via* step (b), into the membrane by a number of methods:

The catalysed support, when in particle or fibre form, can be added directly to an aqueous solution of the proton-conducting polymer electrolyte material; preferably, an essentially aqueous solution as described in EPA 731 520. The use of this aqueous solution permits the direct addition of the pre-formed catalysed support to the polymer solution, and eliminates the undesirable risk of combustion of the organic solvents that are present in the typically-employed polymer solutions. It may then be possible to cast a membrane directly from this mixture according to any of the commonly practised methods in the art, or to employ the mixture to form a composite membrane by applying it to a pre-formed reinforcing substrate material such as a woven or non-woven fibre network of, for example, glass, silica or polymer fibres.

Alternatively, the catalysed support may, when in particle form, itself comprise the binder used to form the fibre network in a composite membrane. Subsequent to the formation of the substrate fibre network or matrix comprising the catalysed support binding an underlying fibre network, the proton-conducting polymer is coated onto the network to produce the final membrane. Alternatively, when in fibre form, the catalysed support may itself form the fibre network prior to coating with a binder.

Preferably, fibres within the substrate are normally randomly orientated in the x and y direction (in-plane) producing a two-dimensional isotropic structure. Additionally, random orientation in the z direction (through-plane) can be introduced with the inclusion of very short fibres, typically lengths of less than or equal to 0.2mm or very fine fibres, typically of diameters less than or equal to $1\mu\text{m}$. The fibres typically have a diameter in the range of from $0.1\mu\text{m}$ to $50\mu\text{m}$, preferably $0.2\mu\text{m}$ to $20\mu\text{m}$ and, more preferably about $9\mu\text{m}$. The fibres typically have lengths in the range of from 0.05mm to 300mm, suitably 0.5mm to 150mm, preferably 1 to 50mm and, more preferably, about 6mm to 20mm.

10

More preferably, the substrate is porous, typically having at least 50%, suitably at least 75%, of the individual pore sizes being greater than $1\mu\text{m}$ in at least one direction, although a porous substrate wherein some of the pores are less than $1\mu\text{m}$ in all directions is within the scope of the invention. Suitably, for applications in fuel cells, the total thickness of the membrane is less than $200\mu\text{m}$ and preferably less than $100\mu\text{m}$.

In a further embodiment, a laminated membrane comprising more than one polymer-containing layer is provided, at least one layer of which is a membrane prepared by the process of the invention. Where a laminated membrane is formed that comprises more than one such membrane layer, each layer may comprise either the same or different types of fibres and porous substrates, and also the same or different types of polymeric material embedded within the porous substrate of each membrane layer. Using such a laminated structure, it is possible, for example, to tailor the properties of the laminate membrane opposed to the anode and cathode sides in the MEA of a proton exchange membrane fuel cell, for example, to improve water management in the fuel cell, or to be able to use lower cost proton-conducting polymers to form a substantial part of the laminate membrane.

The present invention also relates to an MEA and a method for the manufacture thereof, wherein the membrane comprises a polymer composition produced by a process according to the present invention. A further aspect of the present invention relates to a fuel cell and a method for the manufacture thereof, which fuel cell comprises such a membrane.

The present invention will now be described by way of the following non-limiting example(s).

EXAMPLE 1

A 10wt%Pt catalyst supported on silica (one from the Cab-O-Sil range supplied by Cabot Corporation Ltd, Cab-O-Sil Division, Barry Site, Sully Moors Road, Sully, South Glamorgan CF6 2XP and/or the Aerosil range supplied by Degussa AG, Silicas and Chemical Catalysts, Division Applied Technology, Rodenbacher Chausee 4, D-63403 Hanau

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(-Wolfgang) was prepared. The Pt catalyst (0.125g) was then added to an aqueous Nafion[®] solution (75g of a 5wt% solution), prepared as described in patent specifications numbers EPA 731 520 and US 5,716,437. The mixture was then cast to form a membrane which was air-dried. The membrane can either be laminated at above 100°C between anode and cathode to form the MEA, or combined as a layer with additional membrane layers to form a multilayer membrane for subsequent lamination with the electrode layers to form the MEA.

METHOD PROTOCOL FOR EXAMPLE 2

A randomly orientated fibre network made from glass fibres was fabricated by taking chopped glass fibres (Type A20 BC, from Schuller International Group Inc., PO Box 5108, Denver, Co 80217), of a fibre length of 12mm (0.267g), and glass microfibre (Evanite 608 from Evanite Fibre Corporation, Covallis, Oregon, USA) (0.533g) and dispersing and mixing the fibres in demineralised water (500cm³). A non-woven substrate was fabricated from the resulting mixture in a single step process based on the principles of paper-making technology, as a sheet size of 214 cm² in a standard SCA sheet former (AB Lorentzen & Wettre, Box 4, S-163 93 Stockholm, Sweden).

EXAMPLE 2

A 10wt%Pt catalyst supported on silica (as for Example 1) was prepared. The catalyst was then dispersed in an aqueous solution and sprayed onto the surface of the glass fibre web, prepared using a paper maker as described in the Protocol above. The resulting sheet was dried in air at 150°C. The loading of catalyst on the fibre network was 2.1g/m². This produced a platinum loading on the fibre web of 0.21g/m² corresponding to 0.021mg/cm². The web was then filled with aqueous Nafion[®] applied with a brush coating technique and dried at room temperature to produce a membrane. Two similar membranes were then laminated to produce the final membrane of the invention with a total Pt loading of 0.42g/cm². The membrane can either be laminated at above 100°C between anode and cathode to form the MEA, or combined as a layer with additional membrane layers to form a multilayer membrane for subsequent lamination with the electrode layers to form the MEA.

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Johnson Matthey
Technology Centre

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